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**Satellite observations of ozone and chlorine monoxide concentrations during winter 1992–1993 show that in February 1993 chlorine in the lower stratosphere was mostly in chemically reactive forms. Decreases in stratospheric ozone concentration during February and early March 1993 are consistent with chemical destruction by this reactive chlorine. Comparison with changes in the distribution of long-lived chemical and dynamical tracers shows that the observed decrease cannot have been caused solely by dynamical processes.**

THE large ozone loss over Antarctica in late winter<sup>1</sup> is known to be caused by chlorine chemistry<sup>2–4</sup>. Lower-stratospheric chlorine is converted to reactive forms by heterogeneous chemistry on polar stratospheric clouds (PSCs) followed by the action of sunlight<sup>2,5,6</sup>. Enhanced abundances of chlorine monoxide (ClO), the dominant reactive form of chlorine in the daytime stratosphere, were observed during aircraft flights into the Arctic polar vortex in the 1989 and 1992 winters<sup>7–9</sup>. Global measurements by the Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS) showed ClO abundances in the early January 1992 Arctic lower stratosphere comparable to those in the Antarctic during August<sup>10</sup>. Direct observation of Arctic ozone loss is more difficult than in the Antarctic due to greater dynamical variability in the northern polar vortex<sup>11–13</sup>. The stronger Antarctic vortex isolates the air in which ozone destruction occurs, and is much colder than the Arctic vortex<sup>12–14</sup>. Antarctic lower-stratospheric temperatures typically remain low enough for PSC formation for ~4 months, until after the spring equinox—as opposed to a few days to ~2 months in the Arctic<sup>15</sup>, where temperatures are well above the PSC threshold by the spring equinox. Chemical ozone losses in the Arctic should thus be smaller than in the Antarctic, and will be more difficult to detect against the climatological increase in polar ozone during Arctic winter.

To separate dynamical and chemical effects, we examine observations of ozone and ClO on isentropic surfaces (surfaces of constant potential temperature,  $\theta$ ) in relation to long-lived tracers, and to vortex structure as diagnosed from potential vorticity (PV)<sup>16</sup>. MLS observations of ozone and ClO during the 1992–93 Arctic winter, and their relation to the polar vortex, are compared with those in the 1991–92 Arctic and 1992 Antarctic winters, and with Nimbus 7 Limb Infrared Monitor of the Stratosphere (LIMS) ozone during the 1978–79 Arctic winter. Both the LIMS<sup>17,18</sup> and the MLS<sup>19,20</sup> data used here have been extensively examined and are sufficiently reliable for our examination of seasonal trends. The passive tracers nitrous oxide (N<sub>2</sub>O) and methane (CH<sub>4</sub>), which have no significant stratospheric sources and have chemical lifetimes long compared to dynamical timescales, were measured by the UARS Cryogenic Limb Array Etalon Spectrometer (CLAES)<sup>21</sup> during the 1991–92 and 1992–93 winters. Our examination of the CLAES data used here indicates that they are also reliable for examining temporal trends

in and around the polar vortex. Both MLS and CLAES switch between viewing high northern and high southern latitudes every ~36 days.

Analyses of aircraft<sup>22–24</sup> and MLS<sup>10</sup> observations showed decreasing ozone in the Arctic lower stratosphere (~15–19 km) in January 1992; comparison here of MLS-observed ozone changes with expected transport effects supports the view that lower-stratospheric ozone in January 1992 was depleted by chlorine chemistry.

MLS measured enhanced ClO in the 1992–93 Arctic polar vortex beginning in early December 1992; by mid-February 1993, ClO mixing ratios >1 part per billion by volume (p.p.b.v.) were seen throughout the polar region<sup>10</sup> and persisted until late February. Vortex-averaged ozone on the 465 K isentropic surface (referred to as 'ozone at 465 K' from here on) was observed to decrease by ~20% between mid-February and mid-March. Analyses of long-lived tracer data and vortex evolution show that this decrease could not have been caused solely by transport processes at this time. These results imply significant depletion of ozone by chlorine chemistry in the Arctic lower stratospheric vortex between mid-February and mid-March 1993.

## Ozone in the Arctic vortex

MLS viewed high northern latitudes in December to early January, and mid-February to mid-March in 1991–92 and 1992–93. Figure 1 shows synoptic maps of ozone at 465 K (~19 km) in mid-February and mid-March for three Arctic winters (1979 from LIMS, and 1992 and 1993 from MLS), along with contours of Rossby–Ertel potential vorticity (PV) in the region of strong PV gradients. This region is coincident with strong westerly winds and demarcates the polar vortex. During 1979 and 1993, the vortex was considerably eroded by strong warmings between mid-February and mid-March. There were no strong warmings in late winter 1992, and the size of the 1992 vortex barely changed. Ozone at 465 K in 1992 and 1993 was largely confined within the vortex, in contrast to 1979, when PV gradients were much weaker and the vortex therefore less isolated throughout the winter. In fact, tongues of low ozone within the 1979 vortex in February suggest the possibility of intrusion of lower-latitude air into the vortex<sup>25</sup>. Ozone mixing ratios peak in the mid-stratosphere, near ~35 km at high latitudes; thus, downward transport across isentropic surfaces, expected from diabatic cooling in the

winter stratosphere, should increase vortex ozone at 465 K. Ozone in the vortex increased during late winter in 1979 and 1992, as expected in the absence of significant chemical ozone loss. In 1993, however, ozone in the vortex decreased between mid-February and mid-March. Horizontal transport could

decrease vortex ozone because below  $\sim 550$  K ( $\sim 21$  km), there is less ozone at low latitudes than in the polar vortex. However, Fig. 2 shows that the decrease in 1993 vortex ozone was fairly uniform in time, with no obvious signs (within the resolution of the data) of comparatively ozone-poor low-latitude air entering the vortex. Some ozone-rich air was observed being drawn off the vortex edge (for example, on 18 February and 2, 6 and 10 March), which could have decreased the area of high ozone in the vortex, and thus could alter a vortex average.

Vortex-averaged ozone, ClO and vortex minimum temperatures at 465 K for these three years are shown in Fig. 3. Overall, minimum temperatures were lowest in 1992–93 and highest in 1978–79. Although Arctic lower-stratospheric temperatures generally drop below the PSC formation threshold in only a small region of the vortex, in early January 1992<sup>10</sup> and in February 1993 minimum temperatures are located away from the vortex centre, in a region experiencing both sunlight and strong winds; much of the vortex air thus moves through the region of cold temperatures, increasing the extent of PSC processing within the vortex. Unlike in the Antarctic, stratospheric warmings regularly raise temperatures in the Arctic vortex above the PSC formation threshold<sup>11,13</sup>. This occurred with the onset of a strong warming in mid-January 1979, followed by a major warming in mid-

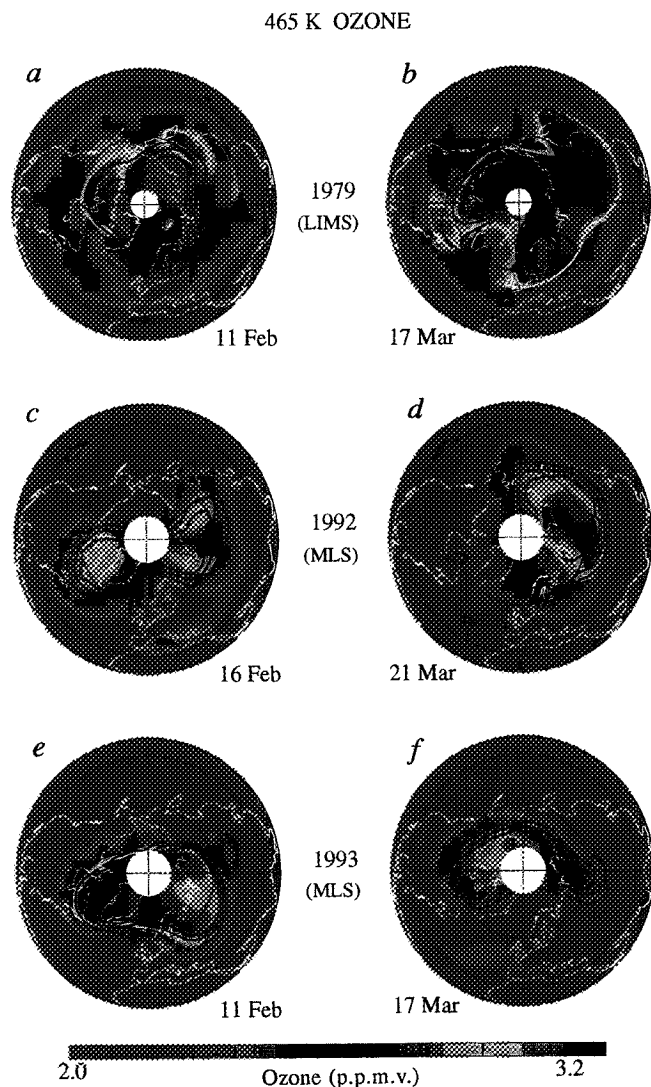


FIG. 1 a–f, Ozone on the 465 K isentropic surface for selected days in mid-February and mid-March 1979, 1992 and 1993. MLS data (c–f) have horizontal resolution of  $\sim 400$  km and vertical resolution of  $\sim 4$  km; the resolution of the LIMS data (a, b) is similar. Precisions (r.m.s.) of individual MLS ozone measurements for this data version (no. 3) are  $\sim 0.2$  p.p.m.v., with absolute accuracies of  $\sim 15$ – $20\%$ . MLS data are gridded for 12:00 GMT using Fourier transform techniques that separate time and longitudinal variations<sup>38</sup>. Potential vorticity (PV) contours of  $2.5$ ,  $3.0$  and  $3.5 \times 10^{-5} \text{ K m}^2 \text{ kg}^{-1} \text{ s}^{-1}$  are overlaid in black. PV is calculated<sup>43</sup> from United Kingdom Meteorological Office (UKMO) assimilation system<sup>39</sup> temperatures and winds for 1992 and 1993, and from LIMS geopotential heights (using a nonlinear primitive equation balance to obtain winds<sup>40</sup>) and temperatures for 1979. Comparison of PV calculated from US National Meteorological Center (NMC) data available from 1978 to the present<sup>41</sup> indicates that the PV fields from different data sets used here are comparable; in particular, the relative strength of the PV gradients is not an artefact of the use of different datasets<sup>42</sup>. UKMO temperatures are used to interpolate gridded MLS data from pressure to isentropic surfaces (MLS temperatures are not currently available at these altitudes). The projection is orthographic, extending to the equator; thin dashed circles show  $30^\circ$  N and  $60^\circ$  N latitudes, and the Greenwich meridian is at the bottom of the plots.

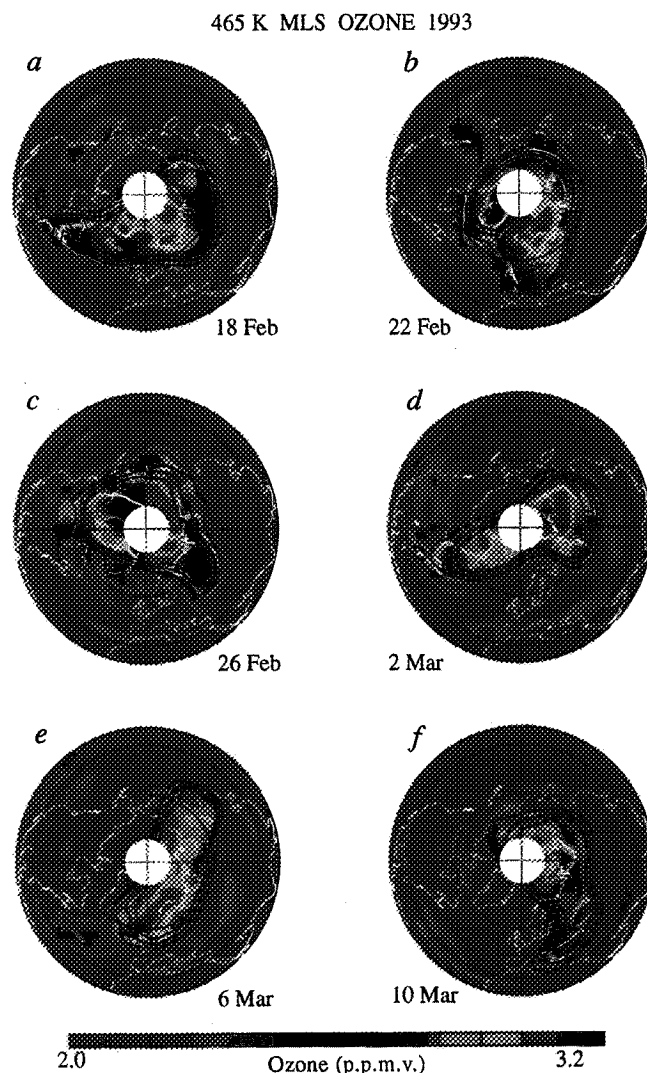


FIG. 2 a–f, MLS ozone on the 465 K isentropic surface at 4-day intervals from 18 February to 10 March 1993. Potential vorticity contours and layout are as in Fig. 1.

February<sup>18</sup>, and with the onset of a strong warming in mid-January 1992<sup>13,26</sup>. The 1993 vortex, however, was relatively strong and cold through mid-February, with strong stratospheric warmings in late February and early March leading to the final warming<sup>27</sup>. The persistence of low temperatures through late February 1993, a period of increasing sunlight at high northern latitudes, implies that high ClO concentrations should be present, which should prolong destruction of ozone by chlorine chemistry in the Arctic vortex during 1993. This is consistent with the MLS observations, as shown below.

The behaviour of LIMS ozone during 1978–79 is included as a baseline for a year when ozone loss in the lower stratosphere due to chlorine chemistry was negligible, because stratospheric chlorine abundances were lower by ~70% than at present. The delayed increase in vortex-averaged ozone in early winter 1978–79 and the brief ozone decrease at the end of January 1979 (Fig.

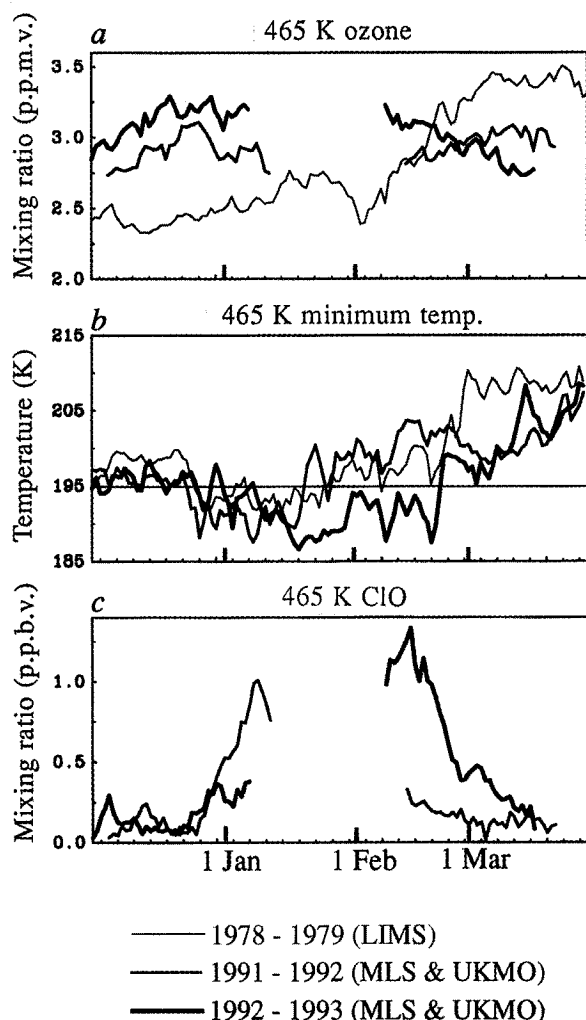
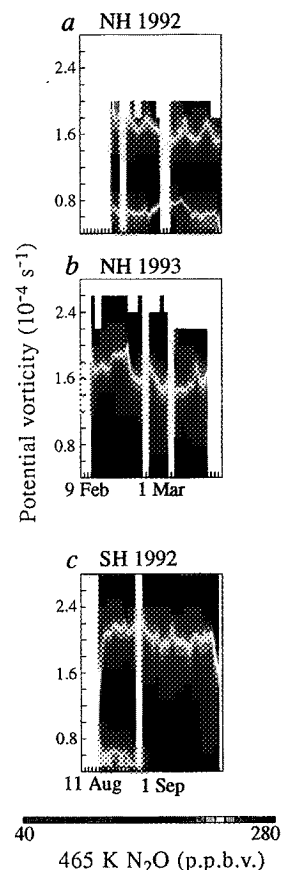


FIG. 3 *a* and *c*, Vortex-averaged mixing ratios, calculated by dividing the area integral of the mixing ratio inside a PV contour (the outermost contour in Figs 1 and 2), by the area enclosed by the contour, of ozone (*a*) and ClO (*c*) on the 465 K isentropic surface, for three Arctic winters; *b*, minimum temperatures (on the same isentropic surface) in the same region used for the vortex averages. Horizontal line in *b* indicates nominal threshold below which PSCs can form. Averaging MLS data as shown here improves the precision to ~0.05 p.p.m.v. for ozone and ~0.1 p.p.b.v. for ClO. The thin red line shows the 1978–79 winter (LIMS data), and the medium green and thick blue lines show the 1991–92 and 1992–93 winters, respectively (MLS ozone and ClO data, and UKMO temperatures). The gaps in MLS data are when it is observing the Southern Hemisphere. Using PV calculated from NMC rather than LIMS or UKMO does not significantly affect any results shown in Figs 3–6.

3*a*) suggest caution when interpreting recent decreases in vortex-averaged ozone as due to chemical rather than dynamical processes. The relatively low vortex ozone at 465 K in December 1978 does not necessarily imply a bias between LIMS and MLS observations, as vortex-averaged ozone was similar (within estimated measurement uncertainties<sup>17,20</sup>) in early November in all three years. However, the dynamical evolution of the vortex was different in 1978–79. The winter observed by LIMS was relatively warm and the lower stratospheric vortex was much weaker, and therefore less isolated, than in either 1991–92 or 1992–93. The behaviour in 1978–79 was not atypical; in fact, a similar pattern, with maximum ozone near the vortex edge, was seen by MLS in the 1993–94 Arctic winter, when the lower stratospheric vortex was also relatively weak. As strong stratospheric warmings perturbed the late January and February 1979 vortex, strongest diabatic descent is expected then at the periphery of the vortex<sup>27</sup>, leading to higher ozone there (for example, Fig. 1*a*). When highest ozone is near the outside edge of the vortex, it is not included in the average shown in Fig. 3*a*. Intrusions of low-latitude, ozone-poor air into the vortex<sup>25</sup> are also more common when it is weak, and rapid changes in LIMS ozone such as the brief decrease in late January 1979 can be due to horizontal transport<sup>18</sup>.

MLS observed enhanced ClO in the Arctic vortex from late December 1991 to mid-January 1992 (Fig. 3*c*), during the time of low temperatures<sup>10</sup> (Fig. 3*b*). By mid-February, when MLS looked north again, ClO had decreased substantially and continued to decay through late February and early March. Vortex-averaged ozone decreased slightly at 465 K from early- to mid-January 1992 (Fig. 3*a*), consistent with vortex-averaged loss

FIG. 4 *a*–*c*, Time series of CLAES N<sub>2</sub>O mixing ratios, averaged around a PV contour, as a function of PV, for 9 February (11 August) to 20 March (19 September) in the Northern Hemisphere, NH (Southern Hemisphere, SH). —PV is used in the Southern Hemisphere, where PV is negative. PV is scaled in 'vorticity units' ( $s^{-1}$ ), where it is divided by a standard atmosphere value of the static stability<sup>13,43</sup>. The bottom of the PV range shown ( $0.4 \times 10^{-4} s^{-1}$ ) is at low latitudes (~20°); PV values of  $\sim(1.0\text{--}1.4) \times 10^{-4} s^{-1}$  are representative of the vortex edge. The CLAES<sup>21</sup> N<sub>2</sub>O measurements have ~400 km horizontal resolution and ~2.5 km vertical resolution<sup>44</sup>. Single profile precision and systematic error estimates in the lower stratosphere for this data version (no. 6) are ~20 p.p.b.v. r.m.s. and ~20%, respectively. N<sub>2</sub>O mixing ratios >~210 p.p.b.v. are less reliable than lower values, due to lack of tangent point sensitivity<sup>44</sup>. On the 465 K isentropic surface shown here, N<sub>2</sub>O values are most reliable poleward of ~50° latitude. The averaging done here results in reliable trends for PV greater than  $\sim(0.6\text{--}0.8) \times 10^{-4} s^{-1}$ . CLAES data are gridded by a simple interpolation of 24 h of data (providing full longitudinal coverage) to a regular latitude–longitude grid. Low N<sub>2</sub>O mixing ratios at low PV in 1992 are artefacts caused by a Pinatubo-aerosol-related problem<sup>44</sup>.



calculated using MLS-observed  $\text{ClO}^{10}$ . The ozone decrease in late January 1979, when ozone destruction by chlorine chemistry was not expected, is comparable to the early January 1992 decrease. But because the vortex was much stronger in 1992 than in 1979, ozone was more confined in 1992, and a comparable decrease caused by dynamics was thus not expected. Furthermore, on a timescale of a month, transport processes should cause an ozone increase during any active period, as seen in February 1979. The fact that vortex-averaged ozone did not increase following the strong warming in mid-January 1992 is circumstantial evidence for chemical ozone loss during that time, consistent with analyses of aircraft data<sup>22-24</sup>.

In the 1992-93 Arctic winter, MLS first detected enhanced  $\text{ClO}$  in early December, and vortex-averaged values at 465 K were  $\sim 0.5$  p.p.b.v. by early January. There were no MLS Arctic observations from mid-January to mid-February 1993, but vortex minimum temperatures remained well below the PSC forma-

tion threshold, suggesting that  $\text{ClO}$  was enhanced throughout this period. When MLS looked north again in February,  $\text{ClO}$  mixing ratios were  $>1$  p.p.b.v. in most of the vortex region. Temperatures remained below the PSC formation threshold until late February, after which  $\text{ClO}$  decreased.

Ozone at 465 K in the 1992-93 Arctic winter is expected to increase more rapidly during the warmings in late February and early March when diabatic descent is enhanced<sup>27</sup>, similar to the

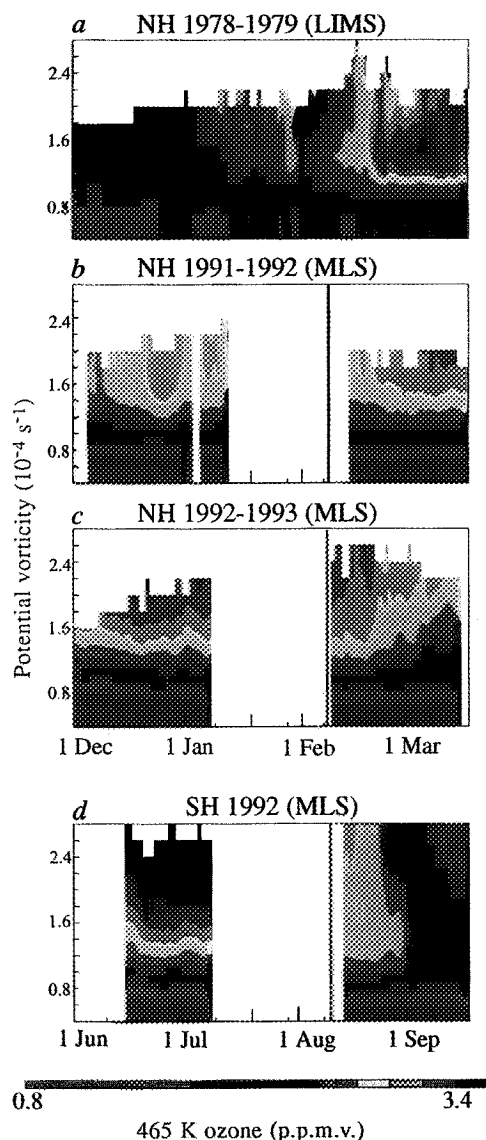


FIG. 5 *a-d*, Time series of ozone mixing ratios averaged around a PV contour, as a function of PV, for 1 December (1 June) to 20 March (19 September) in the Northern Hemisphere, NH (Southern Hemisphere, SH). Heavy vertical black lines indicate the time after which  $\text{N}_2\text{O}$  is shown in Fig. 4. Large gaps in the data are when MLS is looking at the hemisphere not shown.

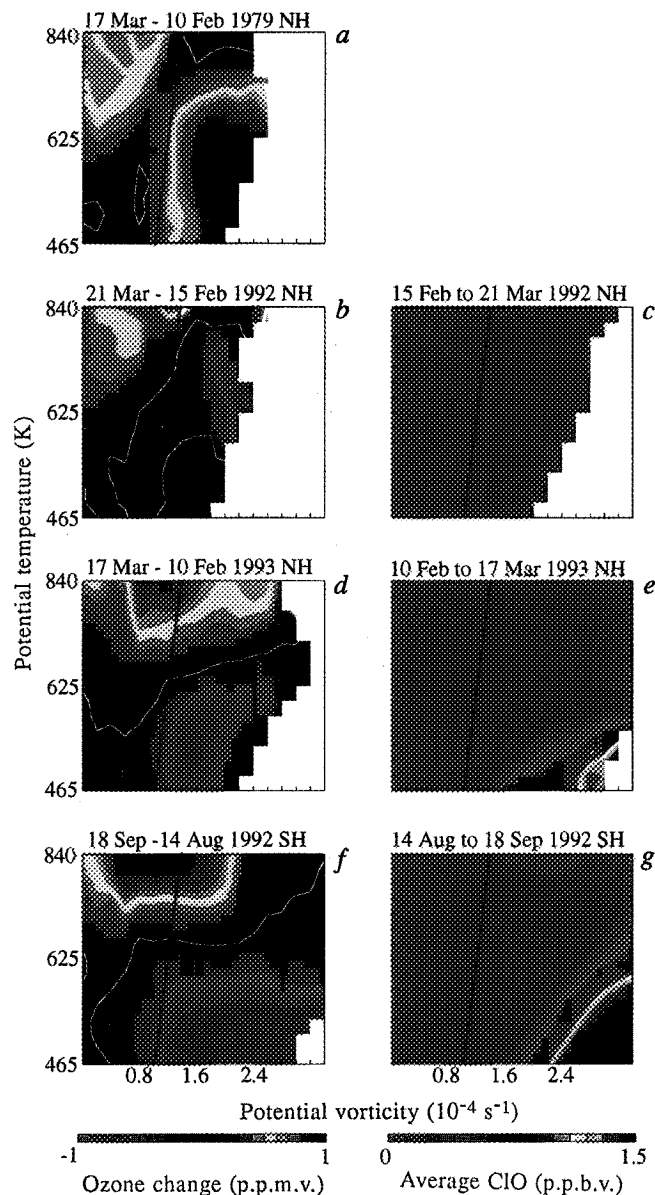


FIG. 6 Change in ozone mixing ratio (left-hand column), and time-averaged  $\text{ClO}$  (right-hand column), over 36 days (the approximate length of time for which MLS observed one hemisphere) in late winter, for three Arctic winters (no  $\text{ClO}$  data exist for 1979) and one Antarctic winter, as a function of PV and potential temperature ( $\theta$ , ref. 31), in the lower to mid-stratosphere. White contour indicates no change. PV is scaled as in Figs 4 and 5, to give a similar range of values throughout the  $\theta$  domain shown. Heavy black lines indicate PV typical of the vortex edge. Because the polar vortex weakens over the time period, especially in the Arctic, maximum PV values are less on the ending than on the beginning day. Ozone difference plots include all PV values present on both days differenced, whereas the  $\text{ClO}$  time-average plots include all PV values present on any day in the period. The  $\text{ClO}$  averaged at the highest PV values thus occurs earlier in the period.

behaviour seen in February 1979. Observed ozone increases gradually in the 1992–93 early winter, as expected. Vortex-averaged ozone is, however, the same in mid-February as in mid-January 1993, and decreases steadily between mid-February and mid-March. Between 10 February and 11 March, the average ozone mixing ratio decreases from approximately 3.2 p.p.m.v. to 2.6 p.p.m.v.,  $\sim 0.7\%$  per day. For comparison, vortex-averaged ozone in the Antarctic in August and September 1992 decreased at  $\sim 1.4\%$  per day<sup>28</sup>. Vortex-averaged ClO mixing ratios in the Antarctic in mid-August 1992 were comparable to mid-February 1993 Arctic values, but the duration of enhanced Arctic ClO was shorter<sup>10,28</sup>, and Antarctic ClO remained enhanced later in the year when more sunlight was present. Calculations using MLS ClO like those done for January 1992<sup>10</sup> give a vortex-averaged ozone loss over the February–March 1993 time period of  $\sim 0.9\%$  per day due to chlorine (and bromine) chemistry. This calculated loss is consistent with the observations, particularly because ozone should increase in absence of chemical depletion.

### Separation of chemical and dynamical effects

The behaviour of ozone is now examined in relation to long-lived tracers<sup>29</sup> to discriminate further between dynamical and chemical contributions to ozone change. As the Arctic vortex is so distorted and variable (Figs 1 and 2), zonal means obscure details of the flow, and average together values from inside and outside the vortex. A more useful average for diagnostic purposes is obtained by changing coordinates from latitude to PV (refs 30–33). Changes in the mixing ratio at constant PV and  $\theta$  provide information on diabatic and chemical processes<sup>29–31,34</sup>. Figure 4 shows time series of CLAES  $N_2O$  on the 465 K isentrope, averaged around PV contours for the Arctic late winters of 1992 and 1993, and the Antarctic late winter of 1992. Similar features are seen in plots of CLAES  $CH_4$ , so only  $N_2O$  is shown here. The  $N_2O$  mixing ratio decreases toward the winter pole and decreases rapidly with height in the lower stratosphere, so diabatic descent would decrease  $N_2O$  along a PV contour on an isentropic surface<sup>34</sup>. The  $N_2O$  shows a slight temporal decrease during February–March 1992 in the vortex at  $PV \geq 1.4 \times 10^{-4} s^{-1}$  (PV scaled in vorticity units is used here—see Fig. 4 caption), indicating weak descent. A steeper decrease is seen in the middle of the February–March 1993 period, as expected from enhanced diabatic descent during stratospheric warmings<sup>27</sup>. The  $N_2O$  level decreases slightly above the  $1.4 \times 10^{-4} s^{-1}$  PV contour during August–September 1992 in the Antarctic, where minor warmings<sup>35,36</sup> are confined to higher altitudes and do not significantly enhance diabatic descent in the lower stratosphere.

Figure 5 shows similar time series for ozone throughout the 1978–79, 1991–92 and 1992–93 Arctic winters, and the 1992 Antarctic winter. Ozone should increase where  $N_2O$  decreases if no chemical depletion occurs. Chemical processes were not expected to be important in the Arctic winter lower stratosphere in 1978–79; the general increase in ozone is consistent with changes expected from diabatic descent. In 1991–92, ozone at 465 K increases slightly in December and in late February, consistent with the transport processes indicated by concurrent changes in  $N_2O$ . In stark contrast, the evolution of ozone in the 1992 Antarctic late winter is very different from that expected from dynamical variations as diagnosed from  $N_2O$ ; ozone decreases rapidly, beginning near the centre of the vortex.

Although less dramatic, the evolution of 465 K ozone during the 1992–93 Arctic winter resembles that in the Antarctic 1992 winter. Arctic ozone slowly increases in December 1992, as does Antarctic ozone in June 1992. But in mid- to late-February 1993, Arctic ozone decreases rapidly well inside the vortex. Comparison with dynamical changes expected from examination of  $N_2O$  (Fig. 4b) indicates that the observed ozone evolution in February–March 1993 is inconsistent with the effects of transport alone. This provides direct observational evidence for chemical depletion of ozone in the Arctic during February–March 1993.

Figure 6 compares the vertical extent of enhanced ClO with ozone changes in late winter. The 1979 ozone (Fig. 6a) shows behaviour expected for no chemical loss during a dynamically active period: a large increase inside the polar vortex throughout the lower stratosphere. Figure 6f and g, for 1992 in the Antarctic, shows the behaviour expected when chemical loss dominates: ozone decreased below  $\sim 650$  K ( $\sim 27$  km) throughout the vortex, in the region of enhanced ClO.

The level of ClO in the Arctic lower stratosphere during February–March 1992 was low compared with that observed when temperatures are below the PSC formation threshold. Ozone in February–March 1992 below  $\sim 650$  K increased slightly inside the vortex, consistent with weak diabatic descent at this time. An ozone decrease of  $\sim 10\%$  occurred in 1992 between 600 and 700 K at highest PV values; a decrease of  $\sim 7\%$  was seen in this region in 1993. First-order estimates of gas-phase ozone destruction reaction rates from catalytic cycles involving  $NO_2$  (from CLAES) and ClO suggest that much of the ozone difference between 1992 and 1993 near 650 K could have been caused by the somewhat larger abundances of these radicals in 1992.

Enhanced Arctic ClO in 1993 extended to near the vortex edge and vertically to  $\sim 580$  K. Although Arctic ClO abundances in mid-February 1993 were comparable to Antarctic values in mid-August 1992, the time average was less in the Arctic because ClO decreased in early March (Fig. 3c), and thus the period of enhancement was less. Ozone decreased in 1993 (Fig. 6d) throughout the vortex below  $\sim 600$  K, in the general region of enhanced ClO.

### Implications for Arctic ozone loss

We have demonstrated above that the  $\sim 20\%$  decrease in Arctic lower-stratospheric ozone in February and March 1993 is inconsistent with changes expected from transport alone during this period. The decrease seems to be consistent with the observed ClO and associated depletion by chlorine chemistry.

Although ozone in the lower stratospheric vortex decreased substantially in late winter 1993, MLS observations showed no significant decrease in column ozone for the same time<sup>20</sup>. Ozone increases rapidly at this time in the Arctic polar mid-stratosphere, compared with much smaller increases in the Antarctic mid-stratosphere<sup>35</sup>. Higher altitudes thus contribute more significantly to the column in the Arctic, masking the lower-stratospheric ozone decrease there. Comparison with a 30-year record of Dobson data<sup>37</sup> shows that the change in Arctic column ozone from MLS between February and March 1993 is less of an increase than commonly observed, but not the smallest increase on record.

Arctic lower-stratospheric temperatures were low enough for PSC formation for  $\sim 1$  month during 1991–92<sup>10,13</sup>, ending in late January. They were low enough for nearly 2 months during 1992–93, until late February, when more sunlight reaches the polar regions and sustains the ozone destruction cycle over larger areas. This year (1993–94), Arctic lower-stratospheric temperatures hovered near and slightly below the PSC formation threshold from December 1993 to late February 1994, when they dropped well below it for  $\sim 2$  weeks; MLS measured substantial enhancement of ClO in the Arctic vortex in late February/early March 1994. The Arctic lower stratosphere in the past 16 years has been sufficiently cold for PSC formation for as long as  $2\frac{1}{2}$  months, and as late as mid-March<sup>15</sup>. It is therefore expected that there will be years in the near future, while stratospheric chlorine levels continue to increase, when ozone depletion in the Arctic lower stratosphere will equal or exceed that in 1993.  $\square$

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1. Farman, J. C., Gardiner, B. G. & Shanklin, J. D. *Nature* **315**, 207–210 (1985).
2. Solomon, S. *Nature* **347**, 347–354 (1990).
3. Anderson, J. G., Brune, W. H. & Proffitt, M. H. *J. geophys. Res.* **94**, 11465–11479 (1989).
4. de Zafra, R. L. et al. *Nature* **328**, 408–411 (1987).
5. Solomon, S., Garcia, R. R., Rowland, F. S. & Wuebbles, D. J. *Nature* **321**, 755–758 (1986).
6. Molina, M. J., Tso, T. L., Molina, L. T. & Wang, F. C. Y. *Science* **238**, 1253–1257 (1987).

7. Brune, W. H., Toohey, D. W., Anderson, J. G. & Chan, K. R. *Geophys. Res. Lett.* **17**, 505–508 (1990).
8. Webster, C. R. et al. *Science* **261**, 1130–1134 (1993).
9. Toohey, D. W. et al. *Science* **261**, 1134–1136 (1993).
10. Waters, J. W. et al. *Nature* **362**, 597–602 (1993).
11. Andrews, D. G. *Pure appl. Geophys.* **130**, 213–232 (1989).
12. Schoeberl, M. R., Lait, L. R., Newman, P. A. & Rosenfield, J. E. *J. geophys. Res.* **97**, 7859–7882 (1992).
13. Manney, G. L. & Zurek, R. W. *Geophys. Res. Lett.* **20**, 1275–1278 (1993).
14. Hartmann, D. L. et al. *J. geophys. Res.* **94**, 16779–16796 (1989).
15. *Northern Hemisphere Winter Summary, 1992–1993, Selected Indicators of Stratospheric Climate* (NOAA, Climate Analysis Centre, National Meteorological Center, Washington DC, 1993).
16. Hoskins, B. J., McIntyre, M. E. & Robertson, A. W. *Q. J. R. met. Soc.* **111**, 877–946 (1985).
17. Remsberg, E. E. et al. *J. geophys. Res.* **89**, 5161–5178 (1984).
18. Leovy, C. B. et al. *J. atmos. Sci.* **42**, 230–244 (1985).
19. Barath, F. T. et al. *J. geophys. Res.* **98**, 10751–10762 (1993).
20. Froidevaux, L. et al. *J. atmos. Sci.* (in the press).
21. Roche, A. E. et al. *J. geophys. Res.* **98**, 10763–10775 (1993).
22. Browell, E. V. et al. *Science* **261**, 1155–1158 (1993).
23. Proffitt, M. H. et al. *Science* **261**, 1150–1154 (1993).
24. Salawitch, R. J. et al. *Science* **261**, 1146–1149 (1993).
25. Plumb, R. A. et al. *J. geophys. Res.* **99**, 1089–1105 (1994).
26. Newman, P. A. et al. *Science* **261**, 1143–1146 (1993).
27. Manney, G. L. et al. *Geophys. Res. Lett.* **21**, 813–816 (1994).
28. Waters, J. W., Froidevaux, L., Manney, G. L., Read, W. G. & Elson, L. S. *Geophys. Res. Lett.* **20**, 1219–1222 (1993).
29. Schoeberl, M. R. et al. *Geophys. Res. Lett.* **17**, 469–472 (1990).
30. Butchart, N. & Remsberg, E. E. *J. atmos. Sci.* **43**, 1319–1339 (1986).
31. Schoeberl, M. R. et al. *J. geophys. Res.* **94**, 16815–16845 (1989).
32. Lait, L. R. et al. *Geophys. Res. Lett.* **17**, 521–524 (1990).
33. Schoeberl, M. R. & Lait, L. R. in *EOS NATO Summer School* (eds Visconti, G. & Gille, J.) 419–431 (North-Holland, Amsterdam, 1992).
34. Haynes, P. H. & McIntyre, M. E. *J. atmos. Sci.* **44**, 828–841 (1987).
35. Manney, G. L. et al. *Geophys. Res. Lett.* **20**, 1279–1282 (1993).
36. Fishbein, E. F. et al. *Geophys. Res. Lett.* **20**, 1255–1258 (1993).
37. *Rep. int. Ozone Trends Panel 1988* (Global Ozone Res. Monitoring Proj. Rep. No. 18, WMO, Geneva, 1988).
38. Elson, L. S. & Froidevaux, L. *J. geophys. Res.* **98**, 23039–23049 (1993).
39. Swinbank, R. & O'Neill, A. *Mon. Weath. Rev.* **122**, 686–702 (1994).
40. Randel, W. J. *J. atmos. Sci.* **44**, 3097–3120 (1987).
41. Finger, F. G. et al. *Bull. Am. met. Soc.* **74**, 789–799 (1993).
42. Manney, G. L., Zurek, R. W., Gelman, M. E., Miller, A. J. & Nagatani, R. *Geophys. Res. Lett.* (submitted).
43. Dunkerton, T. J. & Delisi, D. P. *J. geophys. Res.* **91**, 1199–1208 (1986).
44. Kumer, J. B., Mergenthaler, J. L. & Roche, A. E. *Geophys. Res. Lett.* **20**, 1239–1242 (1993).

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